

Resonant x-ray microdiffraction imaging of polarization switching in ferroelectric $\text{SrBi}_2\text{Ta}_2\text{O}_9$

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Introduction

While much is known about the structure of polarization domains and domain walls in ferroelectric materials at scales as small as the dimensions of chemical unit cell, the evolution of domains in operating devices is in many cases unexplored at length scales orders of magnitude larger. In addition to fundamental interest, there is a pressing need to explore the physics of domain nucleation and growth because domains in ferroelectric materials have a critical role in devices for nonlinear optics, acoustics, and electronics.¹ Operating thin film devices using these materials have typically been studied using electrical or electro-mechanical effects. However, spatially resolved measurements can be complicated by the metal film typically used to form a top electrode. Because the spontaneous polarization in ferroelectric materials is itself a consequence of the materials unit cell symmetry, another approach is to pursue these problems with a direct structural tool. Here we present an x-ray resonant microdiffraction study of polarization reversal in a $\text{SrBi}_2\text{Ta}_2\text{O}_9$ (SBT) thin film in which the polarization switching is imaged with a scanned 0.7 micron diameter x-ray beam.

Methods

An epitaxially grown 250 nm thick SBT film was produced by pulsed laser deposition onto a conducting Nb-doped SrTiO_3 substrate. In order to produce planar devices with a non-zero switchable ferroelectric polarization, a (011) substrate orientation was chosen so that the $\text{SrBi}_2\text{Ta}_2\text{O}_9$ film grew along the [116] direction.² The SBT unit cell lacks a center of inversion symmetry, and thus the intensities of x-ray reflections from regions of the film with different orientations of the ferroelectric **a** axis are not equal; Friedel's law does not apply. At x-ray energies of atomic resonances of the film's component atoms this effect is especially large. In order to estimate the difference in intensity we calculated the structure function of SBT near the Ta L_3 absorption edge as a function of the incident photon energy and the ferroelectric polarization of the film (Fig. 1(a)). It is useful to express this difference as a contrast analogous to the "flipping ratio" used in neutron scattering (Fig. 1(b)). At this resonance, there is a sharp maximum in the contrast between domains with different orientations of the SBT **a** axis relative to the sample surface normal. The structure factor plotted in Fig. 1(b) is calculated from tabulated atomic scattering factors for Sr, Bi and O, and near-resonance optical constants for Ta obtained from absorption measurements on a Ta metal foil. Atomic positions

within the SBT unit cell were taken from powder diffraction measurements by Shimakawa *et al.*³

X-ray microdiffraction was used to spatially resolve the switching of these SBT devices. The radiation from an undulator insertion device at MHATT-CAT was focused using a Fresnel zone plate with a 10 cm focal length. Radiation focused into orders other than the first was rejected using a 20 μm aperture in a Pt-Ir disk. The focal spot size was limited by ambient vibrations to a diameter of 0.7 μm FWHM. The negligible absorption of hard x-rays in thin films is essential for these measurements; there was insignificant ($\sim 10\%$) attenuation of the signal due to absorption in the Pt top electrode.

Results

Figure 2(a) shows images formed by rastering the sample through a $100\text{ }\mu\text{m} \times 100\text{ }\mu\text{m}$ region with the sample goniometer set for SBT {2212} type reflections and the incident beam energy set at 9.884 keV. The two images in Fig. 2(a) were acquired after positive and negative voltage pulses to the top electrode. Fig. 2(b) shows the difference between the images of Fig. 2(a) and reveals the regions in which the remnant polarization remained switched. Only a few regions of the sample show a measurable change in the polarization during the voltage pulse cycle. It is possible that this inhomogeneity is an artifact of the process of fabricating the electrodes of these devices. Images of the same area (not shown) formed with incident beam energy detuned to 20 eV below the Ta L_3 resonance showed no features related to the domain structure. The contrast predicted using Fig. 1(b) reaches 60% while the measurements show a maximum flipping ratio of only 15%. Origins of this discrepancy may include uneven switching within the x-ray spot size and uncertainty associated with the optical constants used to calculate Fig. 1(b).

Discussion

A previous strategy to obtain the response of small areas of ferroelectric devices has been to fabricate devices of small lateral dimensions,⁴ or to apply voltages with a scanned probe.⁵ This microdiffraction technique presents is a new approach to the study of thin film ferroelectric devices and will complement these techniques by allowing ferroelectric devices to be studied simultaneously by structural and electrical probes at the length scale of important switching phenomena.

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FIG. 1. (a) Computed structure factors for $\text{SrBi}_2\text{Ta}_2\text{O}_9$ unit cell orientations resulting from positive and negative pulses. (b) The normalized contrast (defined as $(I_+ - I_-)/(I_+ + I_-)$) for $\{2212\}$ reflections computed using part (a).

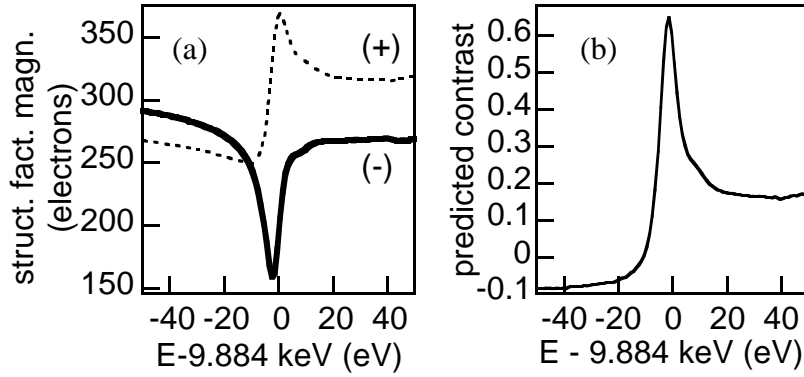


FIG. 2. (a) Images of the intensity of the $\{2212\}$ reflections of the SBT film in a $100\ \mu\text{m} \times 100\ \mu\text{m}$ region following positive and negative polarity voltage pulses of with peak fields of $320\ \text{kV cm}^{-1}$. The ferroelectric capacitor device is formed by a circular Pt contact covering the portion of the image inside the arc from lower left to upper right. (b) The “flipping ratio” for the two images appearing in (a). The area in which the remnant ferroelectric polarization has switched appears as a bright streak through the image.

